Department of Energy
Ohio Field Office
Fernald Area Office
P. O. Box 538705
Cincinnati, Ohio 45253-8705
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DOE-1358-97

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Mr. James A. Saric, Remedial Project Manager
U.S. Environmental Protection Agency
Region V-SRF-5J
77 West Jackson Boulevard
Chicago, Illinois 60604-3590

Mr. Tom Schneider, Project Manager
Ohio Environmental Protection Agency
401 East 5th Street
Dayton, Ohio 45402-2911

Dear Mr. Saric and Mr. Schneider:

RESPONSE TO THE U. S. ENVIRONMENTAL PROTECTION AGENCY AND OHIO ENVIRONMENTAL PROTECTION AGENCY COMMENTS ON THE PROPOSED USE OF THORIUM-232 FINAL REMEDIATION LEVEL AS A BASIS FOR ASSESSING ATTAINMENT OF ALL FINAL REMEDIATION LEVELS IN THE THORIUM SERIES

References: (1) Letter, DOE-0962-97, from J. W. Reising to J. Saric and T. Schneider, "Use of Thorium-232 Final Remediation Level as Basis for Assessing Attainment of All FRLs in the Thorium Decay Series," dated May 19, 1997.

(2) Letter from J. Saric to J. W. Reising, "U. S. EPA Disapproval of the Use of Thorium-232 Final Remediation Level as Basis for Assessing Attainment of all FRLs in the Thorium Series," dated July 29, 1997.

(3) Letter from T. Schneider to J. Reising, "DOE FEMP Disapproval Thorium Progeny FRL Attainment," dated August 12, 1997.

In response to the enclosed References (2) and (3) above, the purpose of this letter is to outline the approach which will be followed to quantify thorium-232, radium-228, and thorium-228 Final Remediation Levels (FRL) within the soils, sediment, and groundwater at the Fernald Environmental Management Project (FEMP). Both the U.S. Environmental Protection Agency (U. S. EPA) and Ohio Environmental Protection Agency (OEPA) agree that given the historical thorium production operations at the FEMP (Reference 1), secular equilibrium in the thorium decay series has been achieved at the FEMP. Nevertheless, however, the OEPA expressed concern (Reference 3) that certain stakeholders may be

expecting to see separate reporting of the FRLs for each of the applicable thorium isotopes. The approach outlined below addresses this concern. Additionally, U. S. EPA expressed concern (Reference 2) that since radium may be more soluble than thorium, a potential environmental dis-equilibrium could be present in sediment and groundwater at the FEMP. As the approach below indicates, the determination of the thorium and radium activity concentrations relies on daughter products which, for instance Actinium-228, have half-lives sufficiently short (Actinium-228 half-life is 6.13 hours) and below radium-228 in the decay chain that concerns of secular equilibrium with respect to radium-228 should not be an issue.

Separately, DOE, Fluor-Daniel Fernald (FDF), and EPA (including both U.S. EPA, OEPA, and Ohio Department of Health) representatives have been discussing, through the real-time radiological work group meetings, the issues surrounding the apparent discrepancies between alpha and gamma spectrometry results from the Area 1, Phase I Certification Program. The approach outlined below should help to address these discrepancies also.

To quantify thorium-232, radium-228 and thorium-228, the FEMP will combine commonly accepted (by the laboratory community) principles with best analytical practices. Some of the decay processes, particularly those involving the radioactive daughters, result in the emission of gamma photons. Five gamma photons are commonly used to quantify thorium-232, radium-228 and thorium-228.

Isotope of Emission	Energy of Emitted Gamma Photon (keV)
Lead-212	238.6
Bismuth-212	727.2
Thallium-208	583.1
Actinium-228	911.1
Actinium-228	969.1

Best analytical practices call for utilizing the information provided by all five of the gamma emitting isotopes to calculate activity concentrations for analytes of interest. Toward that end, the FEMP will calculate an error weighted average to quantify activity concentrations of thorium-232, radium-228, and thorium-228. In the equation below X_1 is the activity concentration of an analyte determined by using the lead-212 gamma photon; X_2 is the activity concentration of an analyte determined by using the bismuth-212 gamma photon; X_3 is the activity concentration of an analyte determined by using the thallium-208 gamma photon; X_4 is the activity concentration of an analyte determined by using the actinium-228 gamma photon (911.1 keV); and X_5 is the activity concentration of an analyte determined by using the actinium-228 gamma photon (969.1 keV). CE_1, CE_2, ... CE_5 are the one sigma counting errors associated with those calculated concentrations.

The error weighted average concentration is then computed as follows (enclosed after the references is a sample calculation from an Area 1, Phase I Certification Unit):

Error Weighted
Average =

$$\frac{X_1(\frac{1}{CE_1})+X_2(\frac{1}{CE_2})+X_3(\frac{1}{CE_3})+X_4(\frac{1}{CE_4})+X_5(\frac{1}{CE_5})}{\frac{1}{CE_1}+\frac{1}{CE_2}+\frac{1}{CE_3}+\frac{1}{CE_4}+\frac{1}{CE_5}}$$

Use of this equation at the on-site laboratory and our contractor laboratories represents best analytical practices for the following reasons:

1) All of the gamma photon information from emitting radioactive daughters is utilized;

2) The gamma photon with the largest measurement error is weighted the least, while the gamma photon having the smallest measurement error is weighted the most; and,

3) The error weighted average will yield a counting error smaller than the counting errors calculated using individual gamma photons (precision improves with more measurements).

If you or your staff should have any questions, please contact Robert Janke at (513) 648-3124.

Sincereiy,

for Johnny W. Reising
Fernald Remedial Action
Project Manager

FEMP:R.J. Janke

Enclosures: As Stated

cc w/enc:

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UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
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K-2113

JUL 29 1997

REPLY TO THE ATTENTION OF: SRF-5J

Mr. Johnny W. Reising
United States Department of Energy
Feed Materials Production Center
P.O. Box 398705
Cincinnati, Ohio 45239-8705

RE: U.S. EPA Disapproval of the "Use of Thorium-232 Final Remediation Level (FRL) as Basis for Assessing Attainment of All FRL in the Thorium Decay Series" Recommendation Document

Dear Mr. Reising:

The United States Environmental Protection Agency (U.S. EPA) has completed its review of the United States Department of Energy's (U.S. DOE) "Use of Thorium-232 Final Remediation Level (FRL) as Basis for Assessing Attainment of All FRL in the Thorium Decay Series" recommendation document. This document, which is dated May 19, 1997, presents U.S. DOE's rationale for using the FRL for thorium-232 to assess the attainment of FRLs in soil, sediment, and groundwater for the thorium-232, thorium-228 and radium-228 members of the thorium decay series. U.S. EPA's review of the document focused on the technical adequacy of U.S. DOE's rationale for using this proposed approach.

Other recent reports, such as U.S. DOE's *Characterization Comparability Study* dated May 1997, have noted significant discrepancies between the results from Th-232 analyses by alpha spectroscopy and those by gamma spectroscopy. This apparent bias must be resolved and result in valid procedures for the analyses of Th-232 in environmental media at Fernald.

U.S. DOE's document inconsistently presents the half-life of radium-228. Table 2 of the attachment identifies the half-life of radium-228 as 6.7 years. Page 4 of the attachment identifies the half-life of radium-228 as 5.75 years. U.S. DOE should consistently present and use the same value for the half-life of radium-228. Because U.S. EPA uses the 5.75 year half-life in determining slope factors and similar risk-based numbers, U.S. EPA recommends that U.S. DOE use this value.

Janke(rj) partial action response to dee-0962-97 (10499) 5

U.S. EPA concurs that sufficient time has elasped since 1972 to allow secular equilibrium to be substantially achieved for any thorium-232 released to soils at the site. However, radium may be more soluble than thorium and could be present at various concentrations in sediment and groundwater. The distribution coefficients and subsequent solubilities for thorium and its progeny should be determined for a wide range of pH conditions and under a wide range of organic matter variations before secular conditions can be assumed. Therefore, U.S. DOE's proposed approach of using the FRL for thorium-232 to assess the attainment of the FRLs for all three radionuclides in groundwater and sediment at the site is not acceptable.

Therefore, U.S. EPA disapproves the "Use of Th-232 Final Remediation Level (FRL) as Basis for Assessing Attainment of All FRL in the Thorium Decay Series" recommendation document. Please contact Gene Jablonowski at (312)886-4591 or myself at (312) 886-0992 if you have any questions regarding this matter.

Sincerely,

James A. Saric
Remedial Project Manager
Federal Facilities Section
SFD Remedial Response Branch #2

cc: Tom Schneider, OEPA-SWDO
Tom Ontko, OEPA-SWDO
Bill Murphie, U.S. DOE-HDQ
John Bradburne, FERMCO
Terry Hagen, FERMCO
Tom Walsh, FERMCO

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George V. Voinovich, Gover
Nancy P. Hollister, Lt. Gover
Donald R. Schregardus, Dire

August 12, 1997

RE: DOE FEMP
DISAPPROVAL
THORIUM PROGENY
FRL ATTAINMENT

Mr. Johnny Reising
U.S. Department of Energy, Fernald Area Office
P.O. Box 538705
Cincinnati, OH 45253-8705

Dear Mr. Reising:

The Ohio Environmental Protection Agency and the Ohio Department of Health-Bureau of Radiation Protection have reviewed your letter "Use of Th-232 FRL as a basis for assessing attainment of all FRLs in the Thorium series." We found the discussion of secular equilibrium between thorium-232 and its progeny and the historical processes involving thorium to be persuasive. We concur that all members of the thorium series are very near to secular equilibrium. However, we are compelled to disapprove the use of thorium as a basis for certifying for its progeny for the following reasons:

1. The Operable Unit 5 Record of Decision and statements made to Stakeholders at public meetings both implied that each contaminant of concern would be individually determined.
2. The only difference between the proposal and the baseline is in the manner that existing data from the various decay processes is statistically manipulated. The proposal under consideration still would require that samples be taken and worked up in the laboratory.
3. Potential savings in time or costs for the new proposal over the standard methods used in Area 1, Phase 1 are minimal.

If you have any questions, please call Tom Ontko or me.

Sincerely,

Tom Ontko

for

Thomas A. Schneider
Fernald Project Manager
Office of Federal Facilities Oversight

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cc: Jim Saric, U.S. EPA
Terry Hagen, FDF
Ruth Vandergrift, ODH
Manager, TPSS/DERR,CO
Dave Ward, HSIGeoTrans
Bob Geiger, PRC

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EXAMPLE CALCULATION OF TH-232 CONCENTRATION USING ERROR - WEIGHTED AVERGE METHODOLOGY

EXAMPLE SAMPLE Calculation	ISOTOPE	GAMMA ENERGY LINE (Kev)	GAMMA ENERGY ABUNDANCIES (%)	RESULT (pCi/g)	1-Sigma COUNTING ERROR (pCi/g)	$\frac{\text{Th-232}}{\text{C.E}}$	$\frac{1}{\text{C.E}}$	$\Sigma\frac{\text{Th-232}}{\text{C.E}}$	$\Sigma\frac{1}{\text{C.E}}$	$\frac{\Sigma\frac{\text{Th-232}}{\text{C.E}}}{\Sigma\frac{1}{\text{C.E}}}$ (pCi/g)
EPI LABORATORY SAMPLE (AREA 1, PHASE I CERTIFICATION)	Pb-212	238.6	44.60	1.32	0.09	14.666	11.111	45.66	39.913	1.14
	Bi-212	727.2	11.80	1.02	0.18	5.666	5.555			
	Tl-208	583.1	84.20*	1.02	0.10	10.200	10.000			
	AC-228	911.1	27.70	1.23	0.13	9.462	7.692			
	AC-228	969.1	16.60	1.02	0.18	5.666	5.555			

*Does not include .359 branching ratio from Bi-212.